Experimental analysis of surface plasmon behavior in metallic circular slits

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Using a focused ion beam, the authors fabricated metallic circular slits onto a glass substrate coated with silver film. The influence of the number of slits and the focusing light phenomena was investigated by capturing the light transmitted through the circular slits. They demonstrate experimentally that the circular grating formed by a set of periodical slits can excite both stronger surface plasmon (SP) and localized surface plasmon as the number of slits increases. They found that the SP tended to congregate at the center of the circular grating, and that the reemitted light could be used to achieve a focusing phenomenon. © *2007 American Institute of Physics*.
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The optical properties of metallic nanostructures have attracted the attention of many scientists. Out of the many interesting properties studied, surface plasmon appears to be one of the most interesting optical phenomenon. According to classical theory,[1] a small aperture has a large cut-off frequency such that light cannot pass through it. Nevertheless, it is known that the aperture can play a role in providing enough of a momentum to convert the propagated light into an evanescent wave and excite the surface plasmon (SP). In 1998, Ebbesen *et al.* identified this extraordinary transmission phenomenon.[2] Ebbesen and co-workers found that a metallic hole array will couple incident light into SP on the incident side such that energy wave will be carried by the SP to be reemitted at the exiting side through the holes.[3,4] Furthermore, the shape of the nanostructure can be used to determine the mode of the SP.[5] In this letter, we will investigate the role of the circular slit number and the focusing phenomenon.

The optical response of a metallic circular slit array has been previously examined theoretically[6] and experimentally.[7,8] A circular slit can be viewed as a coaxial waveguide and the metallic coaxial waveguide can provide a long cut-off wavelength such that the ring array can enhance optical transmission up to 90%.

The shape of the metallic nanostructure that can excite the SP efficiently has been extensively investigated due to its potential to be adopted for creating very useful photonic devices.[9] In comparison, a single metallic circular ring was found to have a very different but interesting optical phenomenon. As the opening width of a circular slit becomes narrower, the optical transmission spectra will be characterized by a redshift phenomenon. Baida and co-workers[10,11] pointed out that the TE_{m1} mode (where the electric field is at a transverse plane) would dominate the optical transmission of the narrow ring such that

$$\lambda_{c,\mathrm{TE}_{m1}} \approx \frac{\pi n(a+b)}{m}, \tag{1}$$

where a and b are the outer and inner radii, and n is the refractive index of the gap. In this letter, we used a focused ion beam (FIB) system (FEI Nova 200) to fabricate the circular slit structure. First, we deposited silver film onto the glass substrate by utilizing a sputtering process, which was followed up by using a FIB to mill the nanostructure directly. We then captured the normal incidence optical spectra by using a confocal microscope equipped with a spectrometer. The signal was detected by a nitrogen-cooled charge-coupled device (Jobin Yvon Triax 320). As the circular slit was of a rotationally symmetric shape, we used nonpolarized collimated light as our illumination source. Figure 1 shows the spectra of the various circular slits at different opening widths and where the outer radii were identical in all cases. The rings were fabricated on the same 235 nm thick silver film deposited onto the glass substrate. It is clear from the

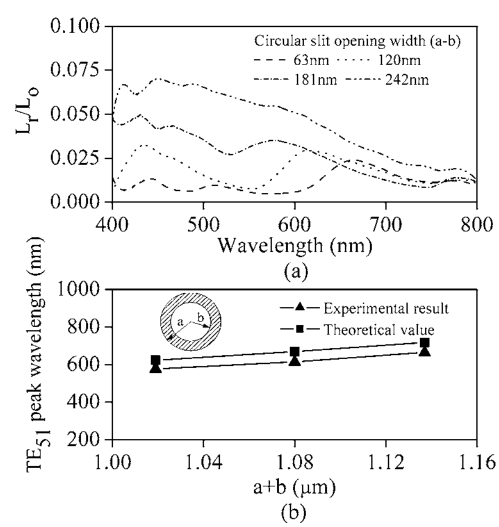

FIG. 1. Optical transmission of a single metallic ring: (a) L_o is the intensity of the halogen lamp light source (outer radius a is fixed at 0.6 μm, and the various line types correspond to the different circular slit opening widths and where Ag film thickness was 235 nm), and (b) comparison of the experimental results with the theoretical values (TE_{51}).

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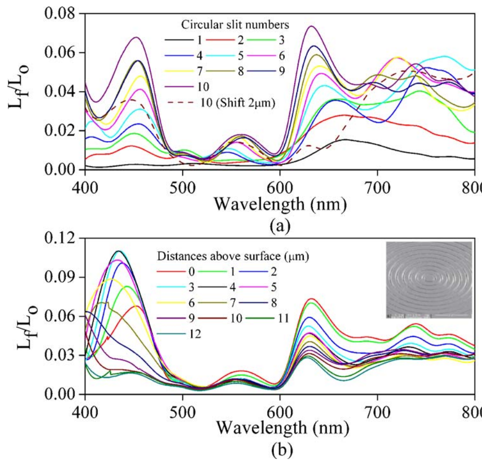

FIG. 2. (Color online) Optical transmission of a circular grating: (a) Ag thickness was 245 nm, with a circular slit width opening of 65 nm at a period of 600 nm and outer radius of first circular slit at 0.6 μm. The various colors represent the various circular slit numbers. The dashed line indicates the transmission spectrum as obtained by shifting the measuring point 2 μm from the center of the grating. (b) The circular grating has ten circular slits (the opening width of the circular slit was 65 nm and Ag film thickness on a glass substrate was 245 nm). The various colors represent the various distances above the metal surface along with the scanning electron microscopy micrograph of the circular grating.

optical transmission curves that different opening widths exert different optical properties. When the opening width is narrow, the transmission spectra will possess a redshift phenomenon. The experimental results agree well with the theoretical predictions [Eq. (1)]. We also found that the TE_{m1} mode does indeed dominate the optical transmission.

In the literature, no prior study discusses the influence of the number of circular slits on the transmission spectra. Figure 2(a) displays the evolution of the transmission spectra as the number of circular slits increases. It shows circular gratings with a different number of circular slits on a 245 nm thick silver film deposited onto the glass substrate and with a circular slit opening width of 65 nm.

Our experimental setup shows a circular grating with ten circular slits showing three main peaks (446, 562, and 632 nm). The intensity at two of these peaks (446 and 632 nm) clearly increases as the number of circular slits gradually increases. It means that these two peaks (446 and 632 nm) are strongly related to the periodicity of the nanostructure and can be attributed to the SP mode. The dashed line in Fig. 2(a) demonstrates the case when the SP is congregated at the center of the circular grating. Since a confocal microscope equipped with a spectrometer is capable of measuring a single point spectrum, we can shift the measuring point to examine the congregating phenomenon of the SP along the transverse direction of the metal surface. When we did this, we found that the intensity of the two 446 and 632 nm peaks could be attributed to the SP as they decreased quickly when we shifted the measurement point 2 μm away from the center of the circular grating. However, the intensity of the other peak (562 nm) only decreased slowly when we shifted the measurement point. We can thus attribute this 562 nm peak to a localized surface plasmon (LSP) mode as its transmission intensity is not as strongly dependent on it position. It is known that a coupled LSP mode relies on the shape of the nanostructure,[12,13] and a single circular slit with a narrower gap has an intrinsically localized electromagnetic mode. The experimental data shown above demonstrated that the circular slit nanostructure can excite both the SP and LSP simultaneously as the number of circular slits increases.

All the above-mentioned transmission spectra shown in Fig. 2(a) were measured directly at the exit surface of the circular slit nanostructures. Figure 2(b) can be used to examine the focusing phenomenon above a metal surface. The beaming phenomenon[14–16] means that the light will propagate along a certain direction in free space. It is intriguingly to explore the focusing phenomenon as the image plane gradually moves away from the exit surface of the circular slit nanostructures. Figure 2(b) displays the focusing phenomenon. We captured the transmission spectra at every single micrometer directly above the metal surface by using a spectrometer and an inverted microscope (Zeiss Axiovert 200). The period of the circular grating obtained was 600 nm and the circular slit opening width obtained was 65 nm. The transmission spectra of this circular grating with ten circular slits were taken at the center point directly above the exit surface of the structure from the metal surface to 12 μm [see Fig. 2(b)]. The transmission spectrum of a circular grating with ten circular slits shows three peaks as seen from a focal plane. The intensity of the 446 nm peak increased as the image plane moved away from the focal plane, and it possessed a different transmission peak at various image planes. This means that the different wavelength light will focus directly above the metal surface. The intensity of a 446 nm peak has a maximum value right directly above the metal surface. This behavior is similar to the focusing phenomenon of a diffracted lens since the intensity of the peak is not at a maximum value at the metal surface. However, the intensity of a 632 nm peak will decay as the image plane moves farther away from the metal surface. It should be noted that the focal length formed is extremely short. Our inverted microscope had a 1 μm resolution and thus the focusing phenomenon could not be easily observed by using this adjusting mechanism. The SP wavelength on the silver film coupled by the 632 nm wavelength incident light wave was about 610 nm, which was close to the period of the circular grating. In the study by Steele *et al.*[17] they concluded that when the period of circular grating approaches in proximity to the SP wavelength, the central intensity of the circular grating achieves its maximum at a near-field region. Extending the conclusion of Steele *et al.* to our work, we can use it to explain why the intensity of our 632 nm peak in Fig. 2(b) was stronger near the surface.

The single circular slit has a distribution of a Fresnel diffraction (F_{ring}) at a far-field region which can be represented as

$$F_{\mathrm{ring}} = \frac{e^{jkz}}{jr}\left(aJ_1\left(ka\frac{r}{z}\right) - bJ_1\left(kb\frac{r}{z}\right)\right), \tag{2}$$

where a and b are the outer and inner radii, respectively, r is the position of the transverse plane, J_1 is the Bessel function of the first kind, z is the propagating length, and k is the wave number. From Eq. (2), it should be pointed out that the energy distribution will be concentrated at the center. The circular grating constructed by many circular slits and its optical energy distribution also will be concentrated at the center, similar to that of a single ring. A different ring will provide a different field distribution, and an interference phenomenon can be observed as the number of circular slits changes. However, as we used a halogen lamp as our light source, the destructive interference could not be observed clearly at the center as the number of circular slits changes because incoherent light did not construct interference very well.

We know that the initial phase of a focused Gaussian beam is curved. Hence, it will be focused at a certain position after propagating in free space. Therefore, if the inner and outer radii become smaller, the cut-off wavelength would be reduced [Eq (1)]. It will be different from a slotted slit which would require altering the slit width to change the cut-off wavelength. When the cut-off wavelength of the slit decreases, the phase velocity of the SP in the slit becomes slower.[18]

If we compare a two-dimensional system with a one-dimensional system, we can see that the light passing every single ring on a circular grating will have a different phase at the exiting side due to different inner and outer radii. Therefore, the phase velocity will be slower at the inner circular slit than at the outer circular slit. From a paraxial approximation, we see that

$$\Delta\phi \approx \frac{\Delta z}{2k}\frac{\nabla_T^2 u}{u}, \quad k\Delta z + \Delta\phi = \text{const}, \tag{3}$$

where $\Delta\phi$ and Δz are the phase difference after propagation and the propagated length, respectively, $\nabla_T^2 u$ and u are the curvature of the phase difference and the phase difference, respectively. When the light passed through the circular slit, we obtained different phases at the exiting side due to different phase velocities in each ring. Thus, phase velocity represents the phase difference at the exiting side. When $\nabla_T^2 u$ is negative; the phase velocity of the propagating wave becomes faster in free space [see Eq. (3)]. As light passes through the circular slit, the curvature of the phase difference near the center will be negative. Since the phase velocity near the center is faster than at any other point in the circular slit, it will focus after propagating in free space.

The phase velocity in a circular slit is related to the sum of the inner and outer radii. The circular slit will allow the light to focus after propagating in free space even if every circular slit has the same opening width. Therefore we can change these parameters to alter the focusing phenomenon of the circular grating. This focusing optical head is similar to a traditional optical lens, and the transmitted light will focus after propagating some distance. The intensity of the transmitted light increases as the number of circular slits increases, and the SP modes will possess propagating behavior along the metal surface in addition to possessing a focusing phenomenon. Thus, we can adopt a SP mode to design a device[19] for nanolithography applications[20] such that it can reduce the required exposure time due to its high transmitted energy and can provide a single point light source to write various geometrical patterns.

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